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Theoretical Potential of the CANDU Core for Transmutation of PWR TRU Nuclides

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1. Introduction

Transuranic (TRU) nuclides generated in spent fuels from the Light Water Reactor (LWR) pose a big challenge in nuclear waste management due to its potential diversion, high radiological toxicity and long lifetime. One of the potential approaches for dealing with spent nuclear fuel is to fully utilize (transmute) the TRU nuclides in nuclear reactors as nuclear fuels.

There have been many studies about utilizing TRU nuclides in CANDU reactors since many design features of CANDU reactor make it uniquely suitable for transmuting TRU nuclides, such as simple fabrication and handling of active fuels, precise management of core reactivity, high neutron economy and so on [1]. The concept of MOX (mixed oxides) fuel, which mixes actinides with natural uranium, and IMF (inert matrix fuel), which mixes actinides with silicon carbide, were proposed previously. The total transuranic element destruction for the IMF (71%) is much higher than in the MOX fuels (40%) [2].

Although the IMF can achieve higher burnup, there is a shortcoming in the structural integrity during high burnup. Since CANDU fuel bundle does not have gas plenum and its cladding is relatively thin, a special fuel design called Fully Ceramic Microencapsulated (FCM) fuel to deal with the TRU transmutation in CANDU was recently proposed [3]. In the FCM fuel, the TRISO fuel particles are randomly dispersed within SiC matrix. The kernel region is filled with $TRUO_2$. SiC is chosen as the matrix material for its tiny dimension change of neutron irradiation and small neutron absorption. Then the TRISO-based FCM pellets are loaded into the Zr alloy tube. Since most of the fission products can be confined within the SiC coating layer of the TRISO particles even at an extremely high burnup, a high burnup can be allowable in the CANDU core. Another important advantage of the FCM fuel is that the fuel temperature is much lower than the temperature in the conventional CANDU core due to the high thermal conductivity of SiC matrix.

In the previous study, a high discharge burnup around 695.13 GWd/MTRU could be achieved by using TRU FCM fuels with 4.3% reactivity loss and 45% TRISO packing fraction [3]. The TRU consumption rate was 71.16%, which was similar to IMF. This paper explores a potential method to increase the incineration of TRU nuclides from PWR for TRISO-based FCM pellets in CANDU reactors by reducing the capture reaction rate of structure material.

2. Analysis Models and Methods

In this work, the analysis is done only for a 2-D fuel assembly lattice because the core characteristics can be well determined through lattice analyses in CANDU core. The lattice calculation is performed for a CANFLEX fuel bundle, as illustrated in Fig. 1, which can improve the operating and safety margin, as well as the economics of a CANDU reactor.

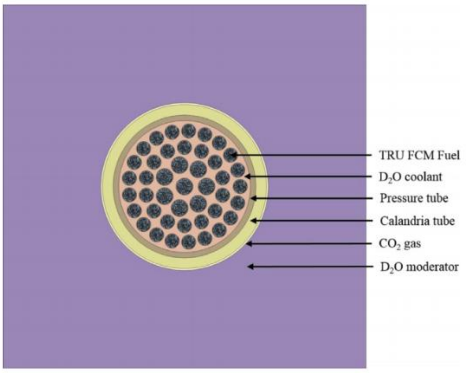


Fig. 1. Unit lattice configuration of the FCM-loaded CANFLEX fuel bundle.

Table I. Dimensions and Calculation conditions of CANFLEX fuel bundle.

CANFLEX		Radius (cm)	Temperature (K)
Center and 1st ring	Fuel pin	0.6350	960.16
	Cladding	0.6710	561.16
2nd and 3rd ring	Fuel pin	0.535	960.16
	Cladding	0.568	561.16
Coolant		5.17915	561.16
Pressure tube		5.61266	561.16
Gas annulus		6.44988	342.16
Calandria tube		6.58954	342.16
Pitch		28.575	342.16

The dimension and calculation conditions of the TRU FCM CANFLEX fuel bundle are listed in Table I. The calculation tool is the Serpent Monte Carlo code [4]. The TRISO particles in the form of FCM fuel are explicitly modelled in the Serpent analysis and their locations are randomly determined. The TRISO particle dimensions are listed in Table II with 45% packing fraction. Table III shows the composition of TRU fuel in this study, which is taken from a 30-year-cooled PWR spent nuclear fuel with a burnup of 45GWd/MTHM. It should be noted that the total fraction of fissile fuel, ^{239}Pu and ^{241}Pu, is around 59% in TRU fuel.

Table II. Design parameters of the TRISO particle.

TRISO	Thickness (μm)	Density (g/cm^3)
Kernel	350	10.4
Buffer layer	100	1.05
Inner PyC layer	35	1.9
SiC layer	35	3.18
Outer PyC layer	40	1.9

Table III. Composition of TRU fuel

TRU	Composition (%)
Np-237	4.69
Pu-238	1.27
Pu-239	56.27
Pu-240	20.11
Pu-241	3.04
Pu-242	3.08
Am-241	9.99
Am-243	0.77
Cm-244	0.06

The infinite multiplication factor is calculated by the Serpent code as a function of burnup with the ENDF/B-VII.0 nuclear data library. In the Monte Carlo calculations, the number of the neutron histories is 100,000 in each Monte Carlo cycle and the total number of cycles is 300 including 100 inactive ones. The standard deviation of the infinite multiplication factor during the depletion is about 16 to 27 pcm. The linear power is 12.94 kW/cm.

3. Analysis Results and Discussions

Naturally occurring zirconium is composed of five isotopes, ^{90}Zr, ^{91}Zr, ^{92}Zr, ^{94}Zr and ^{96}Zr. Only ^{96}Zr is unstable and the half-life of ^{96}Zr is around 2.0E+19 years. Of those natural isotopes, ^{90}Zr is the most common, making up 51.45% of all zirconium. The abundance of ^{91}Zr, ^{92}Zr, ^{94}Zr and ^{96}Zr is 11.22%, 17.15%, 17.38%, and 2.8%, respectively.

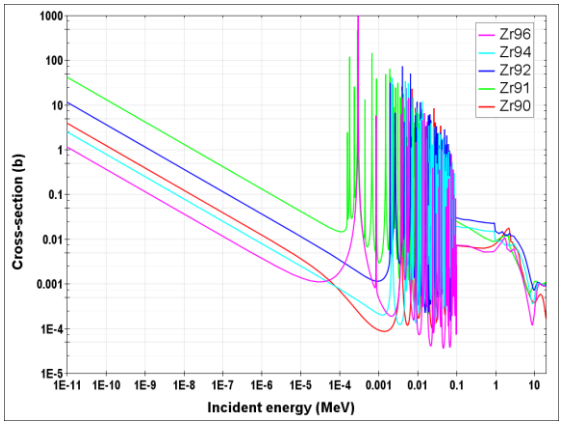


Fig. 2. Capture cross sections of Zr isotopes.

Cross sections of different isotopes of zirconium is shown in Fig. 2. The lowest one is ^{96}Zr, followed by ^{94}Zr and ^{90}Zr. According to their capture cross sections, it is reasonable to use enriched ^{96}Zr in the fabrication of fuel bundle and structural materials. However, because the natural abundance of ^{90}Zr is more than half and the capture cross section is also relatively low, ^{90}Zr is also a competitive choice. In this study, impact of using enriched ^{96}Zr and ^{90}Zr on improving the discharge burnup is investigated. The enriched ^{90}Zr and ^{96}Zr are used in cladding, pressure tube and calandria tube of fuel bundles. The result is compared with that of natural abundance composition.

Fig. 3 shows the evaluation of the infinite multiplication factor values over the fuel burnup for different composition. It indicates that increasing the enrichment of ^{90}Zr or ^{96}Zr results in a slightly higher reactivity or longer fuel lifetime.

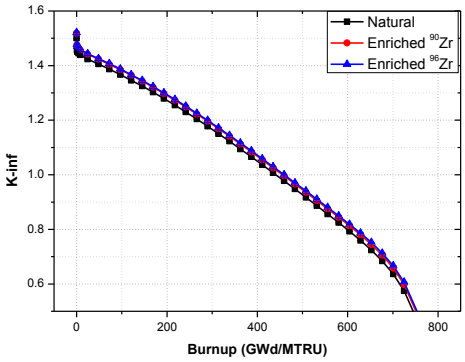


Fig. 3. Infinite multiplication factor with different Zr compositions.

In CANDU lattice analysis, the approximate discharge burnup is determined by the non-linear reactivity model:

$$\int_0^{B_d}(\rho_{(B)} - \rho_{loss})dB = 0 \qquad (1)$$

Where $\rho_{(B)}$ is the lattice reactivity as a function of burnup; ρ_{loss} is the reactivity loss due to the leakage and parasitic capture in CANDU; B_d is the fuel discharge burnup [4].

It is assumed that the reactivity loss is 4.3%, which is coming from the contribution of neutron leakage, neutron absorption by structure geometry and reactivity control device such as adjuster rod. In the current CANDU reactor, the total reactivity worth of the adjuster rods is about 13~15 mk and this positive reactivity is requires to compensate for a large negative reactivity due to xenon buildup around 30 minutes after a reactor trip at full power equilibrium state. However, adjuster rods become not practical nowadays since nuclear reactors cannot be quickly restarted after a sudden reactor trip due to more stringent regulations. And in some Canadian CANDU reactors some or all of the adjuster rods are removed from the core to maximize the fuel utilization. The discharge burnup with a 3% reactivity loss is also calculated, which represents the reactivity loss for a CANDU reactor without adjuster rods. Sixth order polynomial fitting is used to get the function of $\rho_{(B)}$, which is used to calculate discharge burnup as mentioned in equation (1) and the results are

shown in Table IV. From this table, it is clear that increasing the fraction of ^{90}Zr or ^{96}Zr results in a slighter increased discharge burnup due to less neutron absorption cross section of ^{90}Zr or ^{96}Zr.

Table IV Discharge Burnup and of TRU FCM Fuels

Zr composition	Discharge burnup (GWd/MTRU)		
	ρ_{loss}=0	ρ_{loss}=0.03	ρ_{loss}=0.043
Natural	736.21	706.26	691.34
Enriched ^{90}Zr	754.79	726.73	712.75
Enriched ^{96}Zr	766.09	734.73	719.61
Enriched ^{96}Zr in pressure tube and calandria tube	761.85	730.55	715.44

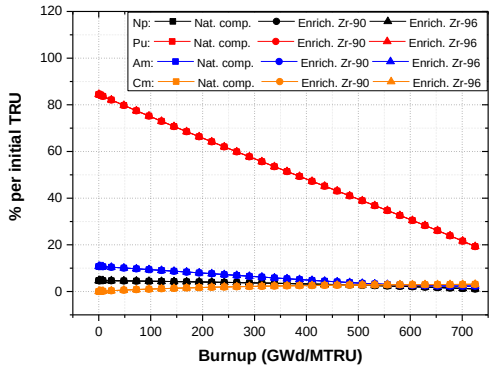


Fig. 4. TRU composition vs burnup.

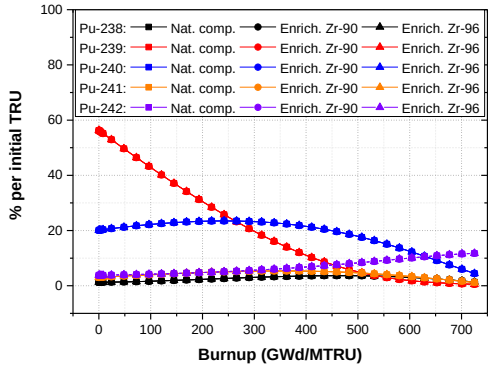
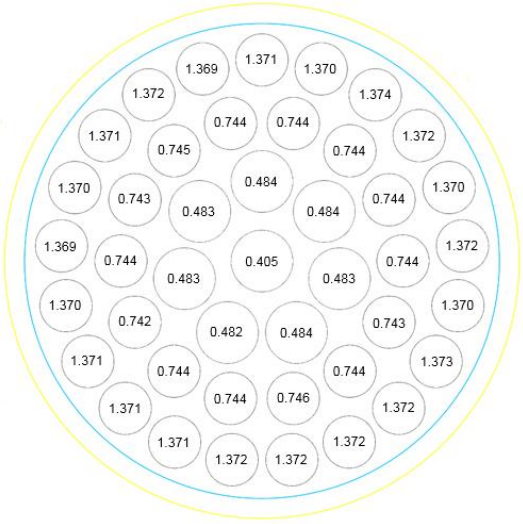


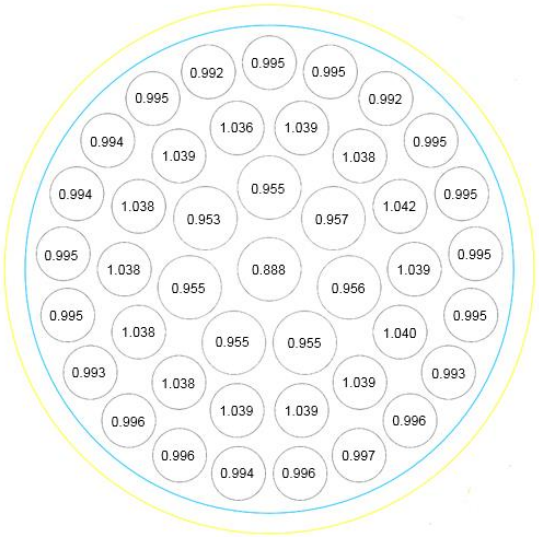
Fig. 5. Pu composition vs burnup.

In Fig. 4, the composition variation with burnup is provided for several transuranic nuclides. This graph shows that transmutation results are similar for the three different zirconium compositions. Plutonium content decreases a lot due to high enrichment at the beginning. Neptunium and americium decrease slowly while curium accumulates during transmutation process.

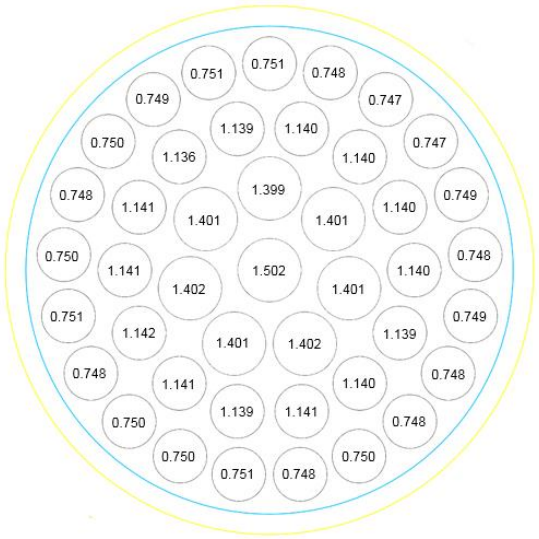
In Fig. 5, the composition variation with burnup is provided for several isotopes of Pu. The transmutation results of different isotopes are also similar for the three different compositions. Pu-239 is almost complete at discharge while Pu-242 increases noticeably due to neutron capture by Pu-241 and the small fission probability of Pu-242 in the thermal spectrum.



a) At zero burnup



b) At mid-burnup



c) At discharge burnup

Fig. 6. Normalized power distribution (natural composition).

Fig. 6 shows the normalized power distribution in the FCM fuel bundles at zero burnup, mid-burnup and discharge burnup condition for natural composition. The trend of power distribution is similar for three different zirconium compositions. In the outer rings, the fission power is the highest at the beginning while it decreases quickly during transmutation process. The fission power in the inner ring has the opposite phenomenon. It is because that the fissile isotopes in outer rings deplete faster than these in inner ring. At the mid-burnup condition, the power profile is relatively flat.

Table V Maximum CVR for TRU FCM fuel

Zr composition	At 0 burnup	At mid burnup
Natural	6.396 mk	10.109 mk
Enriched ^{90}Zr	5.484 mk	8.329 mk
Enriched ^{96}Zr	5.362 mk	8.308 mk
Enriched ^{96}Zr in cladding only	6.467 mk	-
Enriched ^{96}Zr in pressure tube and calandria tube	5.247 mk	-

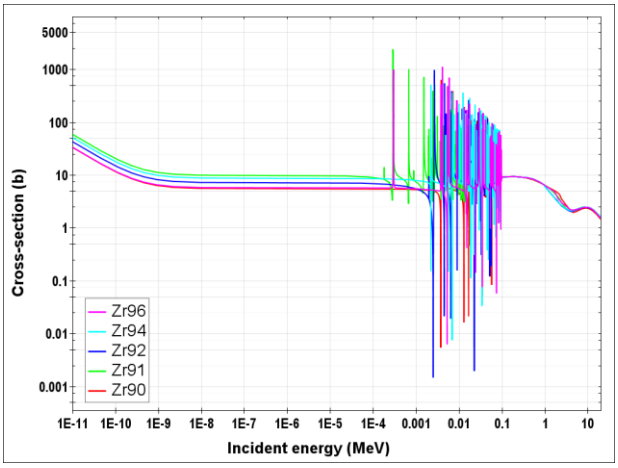


Fig. 7. Elastic scattering cross sections of different isotopes of zirconium.

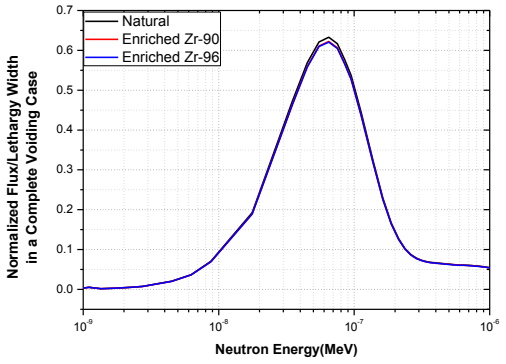


Fig. 8. Neutron spectrum for maximum coolant void.

The maximum CVR (100% voiding) for TRU FCM fuel with natural composition, enriched ^{90}Zr, enriched ^{96}Zr is shown in Table V. It should be noted that the CVR of FCM fuel with enriched zirconium is smaller than that with natural composition, both at zero burnup and middle burnup. It is because that, compared to the other isotopes, ^{90}Zr or ^{96}Zr has a lower elastic scattering cross sections, which is shown in Fig. 7. When crossing calandria tube and pressure tube, neutrons are less thermalized, which will cause reduced thermal neutron flux peak as shown in Fig. 8. On the other hand, if the enriched Zr is used on the cladding only, the CVR is slightly increased. This is because the cladding is really thin. The fully thermalized neutron can easily enter the fuel because the capture cross sections of cladding is now lower.

The enriched Zr-96 when used in cladding, pressure tube and calandria tube will improve the discharge burnup about 4 MWd/MTRU comparing to using it only in pressure tube and calandria tube. In the meanwhile, CVR will decrease around 0.12 mk. Considering the cost of fabricating the cladding, increasing Zr-96 only in pressure tube and calandria tube is a better choice.

4. Conclusions and Future Works

This paper investigates one possible method to increase the discharge burnup of TRU FCM. By increasing the fraction of ^{90}Zr or ^{96}Zr in cladding, pressure tube and calandria tube, higher discharge burnup can be achieved. The composition variation with burnup and the normalized radial power distribution are also analyzed. The coolant void reactivity is smaller with enriched ^{90}Zr or ^{96}Zr in the pressure tube and calandria due to lower elastic scattering cross sections of ^{90}Zr and ^{96}Zr. And increasing Zr-96 only in pressure tube and calandria tube is a better choice.

The current work is based on the ENDF/B-VII.0 nuclear data library. The analysis with new ENDF/B-VII.1 nuclear data library should be performed for a more definitive conclusion. A 3-D analysis should also be considered for the super-deep-burn of TRU nuclides in the CANDU reactor.

Reference

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